Anomalous Shift of Chemical Potential in the Double-Exchange Systems[*]

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Double-exchange system is investigated by the dynamical mean-field theory. We show that the chemical potential shifts as a function of temperature and magnetization, which is anomalously large. We also discuss the influences of dynamic Jahn-Teller effect to the shift of the chemical potential. Measurement of the shift of the chemical potential casts a constraint to theoretical approaches for the magnetoresistance phenomena in $(R,A)\text{MnO}_3$ such as double-exchange effects and dynamic Jahn-Teller effects. We also propose a method to measure the shift of μ.

KEYWORDS: double exchange, perovskite manganite, magnetoresistance

Mechanism of colossal magnetoresistance (MR) in perovskite manganites $(R,A)\text{MnO}_3$ has attracted significant attention. As a canonical model for these compounds, double-exchange (DE) Hamiltonian has been introduced in order to study the ferromagnetism.[1, 2] Recently, interplay between magnetism and transport properties in DE systems are studied in a controlled manner using the dynamical mean-field approach.[3] This model reproduces many experimental results as long as compounds with wide e_g electron bandwidth and high carrier doping, *e.g.* $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ at $x \gtrsim 0.3$, are concerned.

However, compounds with narrower bandwidth show many different properties. One of such examples is that the conductivity shows a crossover from metallic to semiconductive behavior above the Curie temperature T_c by narrowing the electron bandwidth.[4] Dynamical mean-field approach gives temperature insensitive conductivity above T_c and hence fails to explain experimental data in these regions. Several mechanisms to explain semiconductive behavior above T_c have been proposed.

One of such proposals from microscopic points of view is the dynamic Jahn-Teller (JT) theory by Millis *et al.*, where change of electronic states due to interplay between DE and electron-lattice coupling effects is discussed in relation to the MR phenomena.[5, 6] Here semiconductive behavior is considered to be due to formation of lattice polaron with dynamic JT distortion. Experimentally, large lattice distortion in oxygen is observed in $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$.[7, 8, 9] However, direct observation of reconstruction of electronic states in large energy scale with its origin in dynamic lattice distortion has not been reported so far.

Alternatively, effects of Anderson localization in the DE systems with and without charge disorder[10, 11, 12] are also investigated. It is also proposed that long-wavelength spin fluctuation may cause such semiconductive behavior.[13] Apart from the intrinsic bulk nature, magnetic domain boundary effects[14] are also reported as an origin of anomalous MR phenomena.

In this paper, we propose a method to investigate the electronic structure of the DE systems, which casts constraints on possible mechanisms described above for MR in manganites. Namely, the shift of the chemical potential caused by magnetism is anomalously large in the DE systems, and the measurement of the shift shows us informations for the electronic structure of perovskite manganites, such as interplay of charge, spin and lattice degrees of freedom.

We study Zener's DE Hamiltonian,[1]

$$\mathcal{H} = -t \sum_{<ij>,\sigma} \left(c_{i\sigma}^\dagger c_{j\sigma} + h.c. \right) - J_\text{H} \sum_i \vec{S}_i \cdot \vec{\sigma}_i$$
$$-\mu \sum_{i\sigma} n_{i\sigma} - H \sum_i M_i^z. \tag{1}$$

Localized spins $\vec{S}_i$ are treated as classical rotators with the normalization $|\vec{S}| = 1$. Magnetic field is represented by H, and the local magnetization is defined by $M_i^z = (1/2)\sigma_i^z + (3/2)S_i^z$. We apply the dynamical mean-field theory for a system with a semicircular density of states (DOS) with bandwidth W. For Hund's coupling, we choose $J_\text{H} = 4W$ since it reproduce x dependence of T_c as well as MR curve in $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ at $x \sim 0.2$.[3]

In Fig. 1 we show the temperature dependence of the chemical potential at $x = 0.2$ under various magnetic field. At $H = 0$, chemical potential μ is nearly tem-

perature independent above T_c. Below T_c, μ shifts as a function of temperature. In the inset of Fig. 1 we plot μ as a function of magnetic moment $\langle M \rangle^2$. We calculate μ and $\langle M \rangle$, (i) at $H = 0$ by changing temperature in the region $T \leq T_c$, and (ii) at fixed temperature above T_c by changing H. As a result, we see the scaling relation

$$\Delta\mu/W \propto \langle M \rangle^2, \tag{2}$$

where $\Delta\mu \equiv \mu(T, H) - \mu(T = T_c, H = 0)$. We see that $\Delta\mu$ can be as large as $0.1W$.

The origin of the large shift in μ is understood as follows. In Fig. 2 we show the total electron DOS. The width of DOS becomes narrower as temperature becomes higher, with the band center being fixed to the atomic level determined by the energy of e_g orbital and Hund's coupling. Change of the electron bandwidth is understood qualitatively through Anderson-Hasegawa's picture.[2] Electron hopping amplitude is proportional to $\cos(\theta/2)$ where θ is the relative angle of the localized spins. At high temperature, θ deviates from zero due to spin fluctuation, and the amplitude of electron hopping matrix element and hence the bandwidth decreases. This is also shown by the virtual crystal approximation.[15]

Thus, for a fixed band filling, the total change of the DOS width in the entire energy range causes the shift of μ. The change in such a large energy scale controlled by magnetization produce the characteristic feature of the shift of μ in DE systems; namely, the shift of μ as large as a few tenth of W and the scaling relation (2). That the magnetization affects the electronic structure in large energy scale is also seen in the transport phenomena.[3] Since the change of DOS takes place not only in the vicinity of the Fermi surface, we speculate that the present result is not sensitive to the shape of DOS.

The bandwidth of e_g electrons in $La_{1-x}Sr_xMnO_3$ is estimated to be in the order of $W \sim 1eV$ or larger by experiments[14, 16, 17] and first-principle calculations.[18] Similar value for the bandwidth is also estimated from spin wave excitation spectrum[19] as well as T_c for various values of doping[3] by fitting the experimental values with the results for DE Hamiltonian. Then, the shift of μ is estimated to be as large as $0.1eV$.

Let us now consider the case of strong JT coupling. It has been shown that interplay between JT lattice displacements and DE effects produce a reconstruction of electron DOS as a function of temperature and magnetization.[6] When the energy scale of JT distortion potential is comparable to or larger than the electron bandwidth, transport properties of DE systems above T_c will be relevantly affected by the dynamic JT effect, which is one of the possible explanations for MR phenomena in manganites.[5] In this case, temperature dependence of JT distortion amplitude will cause the change in energy split of e_g orbitals in the order of electron bandwidth.
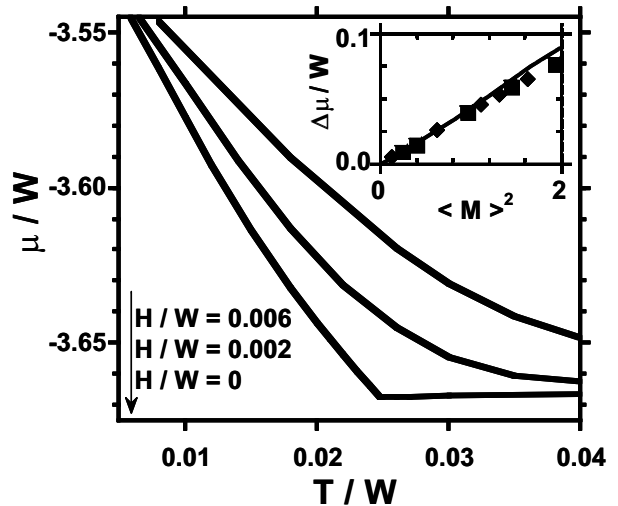

Fig. 1. Temperature dependence of μ at $J_H/W = 4$ and $x = 0.20$ under various magnetic field. Inset: $\Delta\mu/W$ as a function of $\langle M \rangle^2$. Lines show the result for $H = 0$. Squares and diamonds are data at $T = 1.01T_c$ and $1.2T_c$, respectively.
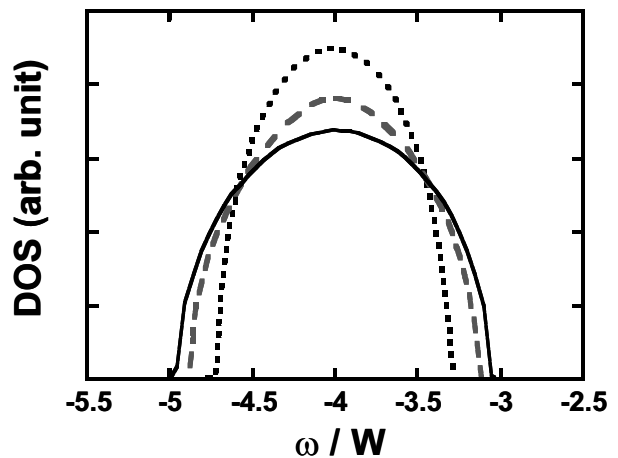

Fig. 2. Temperature dependence of the DOS at $J_H/W = 4$ and $x = 0.20$ obtained by dynamical mean-field approach. Solid, dashed and dotted curves are for $T/T_c = 0.2, 0.5$ and 1.05, respectively. Width of DOS at the paramagnetic phase becomes $1/\sqrt{2}$ times narrower than that for the ground state in the limit $J_H/W \to \infty$.

Therefore, large shift of μ is expected also at above T_c.

Thus, measurement of the shift of μ in $(R,A)MnO_3$ will be one of the crucial tests for the DE mechanism as well as the dynamic JT effect in these materials. Especially, by controlling the bandwidth, we may obtain interplays between charge and lattice distortions through a simultaneous comparison with other experiments such as transport and lattice distortion measurements.

Experimentally, shift of the chemical potential should be observed directly by photoemission provided its en-

ergy resolution is high enough. Here we propose an alternative method to detect the shift of μ. Let us consider a junction of a DE material and a semiconductor. When the Fermi level of the semiconductor is in between those of the DE material at zero and saturated magnetization, which may be realized by bias voltage, carriers in the semiconductor are injected or depleted depending on the magnetic state of the DE material. Namely, conductivity of the semiconductor as well as its Schottky level can be controlled by the magnetism of the DE material. Advantage of this method is that detecting currents do not path through the junction. Disadvantage of this method is, however, that pinning of Fermi level may occur if the junction is not ideal enough, and then the shift may not be detected sensitively.

To summarize, we have shown the large shift of the chemical potential in the DE systems, which is scaled by magnetization. Influences by strong JT coupling is also discussed, and through the measurement of μ in $(R,A)\mathrm{MnO_3}$ we may observe the relevance of dynamic JT effects to its electronic states. A method to measure the shift of μ is proposed.

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